

10/526949 #2

Rec'd PCT/PTO 08 MAR 2005

REC'D 24 NOV 2003

WIPO PCT

P1 1095914

THE UNITED STATES OF AMERICA

TO ALL TO WHOM THESE PRESENTS SHALL COME:

UNITED STATES DEPARTMENT OF COMMERCE
United States Patent and Trademark Office

November 19, 2003

THIS IS TO CERTIFY THAT ANNEXED HERETO IS A TRUE COPY FROM THE RECORDS OF THE UNITED STATES PATENT AND TRADEMARK OFFICE OF THOSE PAPERS OF THE BELOW IDENTIFIED PATENT APPLICATION THAT MET THE REQUIREMENTS TO BE GRANTED A FILING DATE.

APPLICATION NUMBER: 60/415,987

FILING DATE: October 04, 2002

RELATED PCT APPLICATION NUMBER: PCT/US03/31465

By Authority of the
COMMISSIONER OF PATENTS AND TRADEMARKS



N. Woodson
N. WOODSON
Certifying Officer

**PRIORITY
DOCUMENT**

SUBMITTED OR TRANSMITTED IN
COMPLIANCE WITH RULE 17.1(a) OR
(b)

BEST AVAILABLE COPY

10/04/02
J1044 U.S. PTO

10/4/02

Approved for use through 10/31/2002. OMB 0851-0032
U.S. Patent and Trademark Office; U.S. DEPARTMENT OF COMMERCE
PTO/SB/16 (10-01)
the Paperwork Reduction Act of 1995, no persons are required to respond to a collection of information unless it displays a valid OMB control number

PROVISIONAL APPLICATION FOR PATENT COVER SHEET

This is a request for filing a PROVISIONAL APPLICATION FOR PATENT under 37 CFR 1.53(c).

Express Mail Label No.

EL434047691US

| INVENTOR(S) | | | | | |
|---|--|--|--|---|--------------------|
| Given Name (first and middle [if any]) | | Family Name or Surname | | Residence (City and either State or Foreign Country) | |
| J. Keith | | Nelson | | Niskayuna, NY | |
| <input type="checkbox"/> Additional inventors are being named on the _____ separately numbered sheets attached hereto | | | | | |
| TITLE OF THE INVENTION (500 characters max) | | | | | |
| Nanometric Composites as Improved Dielectric Structures | | | | | |
| Direct all correspondence to: CORRESPONDENCE ADDRESS | | | | | |
| <input type="checkbox"/> Customer Number | | Type Customer Number here | | Place Customer Number Bar Code Label here | |
| OR | | | | | |
| <input checked="" type="checkbox"/> Firm or Individual Name | | Rensselaer Polytechnic Institute | | | |
| Address | | Attn: Office of Technology Commercialization | | | |
| Address | | 110 8th Street | | | |
| City | | Troy | State | NY | ZIP 12180 |
| Country | | USA | Telephone | (518) 276-6023 | Fax (518) 276-6380 |
| ENCLOSED APPLICATION PARTS (check all that apply) | | | | | |
| <input checked="" type="checkbox"/> Specification Number of Pages | | 9 | <input type="checkbox"/> CD(s), Number | | |
| <input type="checkbox"/> Drawing(s) Number of Sheets | | | <input type="checkbox"/> Other (specify) | | |
| <input type="checkbox"/> Application Data Sheet. See 37 CFR 1.76 | | | | | |
| METHOD OF PAYMENT OF FILING FEES FOR THIS PROVISIONAL APPLICATION FOR PATENT | | | | | |
| <input checked="" type="checkbox"/> Applicant claims small entity status. See 37 CFR 1.27. | | | | FILING FEE AMOUNT (\$) | |
| <input checked="" type="checkbox"/> A check or money order is enclosed to cover the filing fees | | | | \$80.00 | |
| <input type="checkbox"/> The Commissioner is hereby authorized to charge filing fees or credit any overpayment to Deposit Account Number: | | | | | |
| <input type="checkbox"/> Payment by credit card. Form PTO-2038 is attached. | | | | | |
| The invention was made by an agency of the United States Government or under a contract with an agency of the United States Government. | | | | | |
| <input checked="" type="checkbox"/> No | | | | | |
| <input type="checkbox"/> Yes, the name of the U.S. Government agency and the Government contract number are _____ | | | | | |

Respectfully submitted

SIGNATURE

Charles F. Rancourt

TYPED or PRINTED NAME Charles F. Rancourt

TELEPHONE (518) 276-6023

Date 10/4/02

REGISTRATION NO.
(if appropriate)
Docket Number:

RPI-746

USE ONLY FOR FILING A PROVISIONAL APPLICATION FOR PATENT

This collection of information is required by 37 CFR 1.51. The information is used by the public to file (and by the PTO to process) a provisional application. Confidentiality is governed by 35 U.S.C. 122 and 37 CFR 1.14. This collection is estimated to take 8 hours to complete, including gathering, preparing, and submitting the complete provisional application to the PTO. Time will vary depending upon the individual case. Any comments on the amount of time you require to complete this form and/or suggestions for reducing this burden, should be sent to the Chief Information Officer, U.S. Patent and Trademark Office, U.S. Department of Commerce, Washington, D.C. 20231. DO NOT SEND FEES OR COMPLETED FORMS TO THIS ADDRESS. SEND TO: Box Provisional Application, Assistant Commissioner for Patents, Washington, D.C. 20231.

Under the Paperwork Reduction Act of 1995, no persons are required to respond to a collection of information unless it contains a valid OMB control number

Certificate of Mailing under 37 CFR 1.8

I hereby certify that this correspondence is being deposited with the United States Postal Service with sufficient postage as first class mail in an envelope addressed to:

Assistant Commissioner for Patents
Washington, D.C. 20231

on October 4, 2002
Date

Deborah Lopez-Rouse
Signature

Deborah Lopez-Rouse

Typed or printed name of person of signing Certificate

Note: Each paper must have its own certificate of mailing, or this certificate must identify each submitted paper.

Burden Hour Statement: This form is estimated to take 0.03 hours to complete. Time will vary depending upon the needs of the individual case. Any comments on the amount of time you are required to complete this form should be sent to the Chief Information Officer, U S Patent and Trademark Office, Washington, DC 20231. **DO NOT SEND FEES OR COMPLETED FORMS TO THIS ADDRESS. SEND TO:** Assistant Commissioner for Patents, Washington, DC 20231



Rensselaer

OFFICE OF TECHNOLOGY
COMMERCIALIZATION

Invention Disclosure Form

This form must be completed in its entirety and returned to the Office of Technology Commercialization in J-Building Room 3208 or lopezd3@rpi.edu.

| | | |
|---|-------------------------|------------------|
| 1. Title of Invention: | | |
| Nanometric Composites as Improved Dielectric Structures | | |
| 2. Inventor(s): | Position | Department/Phone |
| J. Keith Nelson | Professor | ECSE/6328 |
| | | |
| | | |
| | | |
| | | |
| | | |
| 3. List dates for the following: | | |
| Invention conceived by inventor (s) | February 2001 | |
| First sketch of Invention prepared | | |
| First written description prepared | May 2002 | |
| First model or full size device completed | | |
| First test or operation of Invention | November 2002 (planned) | |
| First offer of sale (Provide details in #6) | | |
| First sale | | |
| First public disclosure | October 2002 (planned) | |
| 4. The Invention was made under one or more of the following circumstances | | |
| <input type="checkbox"/> In connection with sponsored research at Rensselaer. If so, please identify the contract and if possible, attach a copy. | | |
| <input checked="" type="checkbox"/> In connection with any agreement of any type, which gives, any party rights whatsoever in the Invention. | | |
| <input type="checkbox"/> On my own time (Please provide details) | | |
| 5. List names of each person (other than co-inventors) who have knowledge of Invention and how it was disclosed. | | |
| Professor J.C. Fothergill - University of Leicester, UK | | |
| Professor L.A. Dissardo - University of Leicester, UK | | |
| Collaborators while on sabbatical leave in the UK. | | |

Rensselaer Polytechnic Institute
110 8th Street | Troy, NY 12180-3590 | J-Building
Phone (518) 276-6023 | Fax (518) 276-6380 | www.rpi.edu/dept/otc

RPI - Confidential



Rensselaer

OFFICE OF TECHNOLOGY
COMMERCIALIZATION

6. List all publications, presentations or offers for sale that have been made, which include or describe this Invention.
J.K. Nelson, J.C. Fothergill, L.A. Dissardo & W. Peasgood, "Towards an Understanding of Nanometric Dielectrics", Ann. Rep. IEEE Conf. on Elect. Insul. & Dielect. Phen., 2002 (to appear) - attached.
7. List all known patents, publications and products which include or describe an Invention or technology similar to the Invention.
A. Goel et.al., "Diamond-like Nanocomposite Coatings Possessing High Dielectric Strength", 5th Int. Conf on Cond and Break in Solid Dielectrics", IEEE, 1995, pp. 690-95.
P.O. Henk, T.W. Kortsen & T. Kvarts, "Increasing the Electrical Discharge Endurance of DGEBA Epoxy Resin by the Dispersion of Nanoparticle Silica", High Perform Polym, 11, 1999, pp. 281-96.
8. Provide a description of the Invention along with supporting drawings, reports, photos, etc. The description should at least (1) distinguish the Invention over prior art, (2) identify the advantages of the Invention including all problems solved, and (3) list all alternative methods, materials or competing technologies which would accomplish the same results as the Invention.

Polymers of many types are commonly used as electrical insulation. In many instances, there is need to fill the polymers with materials to modify properties or to reduce the cost. In general, when this is done, the electric strength and other electrical properties are modified in a negative way.

Extensive work carried out by the inventor (using facilities at the University of Leicester, UK and partially supported by the UK EPSRC) has shown that when the filled material is dispersed in the form of nanoparticulates, the properties can be adjusted in a very positive way. It has been shown that when the filler material is reduced in physical size to dimensions of the same order as the polymer chain length, then the particles behave cooperatively with the host matrix. This is in contrast to conventional materials where the fillers have been shown to act as foreign bodies displaying interfacial properties.

In particular, the Maxwell-Wagner process associated with conventional fillers appears to be mitigated by the use of nanofillers. The attendant reduction in interfacial polarization has been demonstrated by the use of a pulsed electroacoustic technique capable of examining the charge (and hence the electric field) profile within the bulk of the insulating structure. An example is provided in Attachment 1 which shows an epoxy thermoset containing 10% inorganic oxide filler (TiO_2) with both micro and nano dimensions. It is evident that the internal fields for the new formulation are nearly a factor of 10 lower than for conventional (micro) material.

The finding is reproducible, and is mirrored in other methods of characterization undertaken:

- Thermally stimulated current analysis
- Dielectric spectroscopy
- Differential scanning calorimetry

The large changes in internal field that can be obtained strongly suggest that new nanocomposite materials can be engineered that have enhanced electric strength and improved voltage endurance properties. Preliminary dielectric strength tests undertaken in 2001 by the discloser on a LDPE / Al_2O_3 system shows this to be so. Comprehensive tests are planned for the fall of 2002 to optimize and verify this using discretionary funding in the first instance.

Rensselaer Polytechnic Institute
110 8th Street | Troy, NY 12180-3590 | J-Building
Phone (518) 276-6023 | Fax (518) 276-6380 | www.rpi.edu/dept/otc

RPI - Confidential



Rensselaer

OFFICE OF TECHNOLOGY
COMMERCIALIZATION

9. Keywords associated with this Invention.

dielectrics, nanotechnology, polymer composite, space charge

10. Provide a status of technology commercialization for the Invention. The description should at least (1) identify all possible uses for the Invention, (2) identify all companies that would be able to communicate the Invention, (3) best method to commercialize the Invention, and (4) indicate the status of work to be done prior to commercialization.

Electrical insulation is a pervasive technology which is a huge commercial business ranging from the thin films used in the microelectronics industry to the large amounts of material used to insulate high-voltage equipment in the power segment of the market. In most instances, the dielectric properties of the insulating structure limits the design. A, say, 20% improvement in performance would thus have significant industrial significance and so the substantial changes that are indicated by this disclosure are believed to be commercially important.

The availability of nanoparticles of a wide range of inorganic oxides offers the possibility of creating a range of new materials with tailored properties and benefits eg. variation in relative permittivity and linearity. Furthermore, it is anticipated that the use of smaller molecules as synthetic additives, chemical coupling agents, triblock copolymers, etc. may permit an element of self assembly of these structures, and create a class of "smart" materials based on nanocomposites to provide auto stress relief and other forms of self compensation.

Towards an understanding of nanometric dielectrics

J. Keith Nelson[§], John C. Fothergill, L.A. Dissado and W. Peasgood

Department of Engineering, University of Leicester, UK

[§]on leave from Rensselaer Polytechnic Institute, Troy, NY, USA

Abstract: Dielectric studies are described aimed at providing an understanding of the charge storage and transport of an epoxy resin containing TiO₂ nanoparticles. Comparative results for conventionally filled composites are given, and the results discussed in terms of the underlying physics. It is shown that nanometric fillers mitigate the interfacial polarization characteristic of conventional materials with a reduction in the internal field accumulations.

Background and Vision

Nanoparticles are the fundamental building blocks in the design and creation of assembled nano-grained larger scale structures with excellent compositional and interfacial flexibility. However, rather surprisingly, the current push to develop nanomaterials based on nanotechnology has not focused much on the opportunities for dielectric materials, but rather centred on optical and mechanical applications. Nonetheless, the few examples in the literature provide encouragement that this is likely to be fertile ground. Furthermore there are good theoretical reasons why the pursuit of nanomaterials for dielectric applications may have particular promise. Some of these have been reviewed by Lewis[1] and by Frechette[2]. While the technology is in its infancy, one may speculate that it may be possible to self-assemble nanodielectrics by providing chemical structures with "hooks" which provide preferential attachment points for the nanostructured materials allowing automatic and predictable self assembly.

Fundamental Issues and Rationale

The use of conventional fillers for polymer materials is well known and is usually employed to reduce the cost of a material or to modify one of the material properties for a particular application. Examples of that would be discharge resistance, thermal expansion, etc. Often the use of such fillers will affect dielectric strength and loss in a negative way. In this context, it is thought that fundamental to controlling the dielectric strength of insulating polymers is the cohesive energy[3] density and the associated free volume[4] of a polymer structure. This may be gauged by examining the

changes in electric strength (up to a factor of 10) exhibited by most polymers as they are taken through their glass transition temperature[5]. In the simplest situation, the bonding of a polymer to a filler can be expected to give a layer of "immobilized" polymer. The size of this layer is critical to the global properties (electrical, mechanical and thermal) of the composite.

Such a picture is not, however, complete since the in-filled material will give rise to space-charge accumulation and the associated Maxwell-Wagner polarization due to the implanted interfaces. Furthermore, the macroscopic theories of interfacial polarization do not incorporate a molecular approach since the response is given by relaxation equations if the wavelength is large in comparison with molecular dimensions. In considering pre-breakdown high-field conduction in pure materials, the existence of localized states within the energy band gap (close to the conduction or valence bands) is usually invoked, so giving rise to a mobility edge for electron (or hole) transport[6]. These states are essentially localized on individual molecules. This is because, unlike the strong covalent bonds of elemental crystalline solids, the intermolecular binding arises from weak van der Waals' forces that do not allow inter-molecular electronic exchange

System Characterization

In order to provide the basis for engineering nanodielectrics, this study has provided a characterization of micro- and nano-particulates of Titanium Dioxide (TiO₂) when embedded in a resin matrix. A Bisphenol-A epoxy (Vantico CY1300 + HY956) was chosen because it was benign (i.e. without other fillers or dilutents), with a low initial viscosity,

| Material + Filler | Size (nm) | Loading (%) | T _g (°C) |
|---------------------------|--------------|-------------|---------------------|
| CY1300 Resin | N/A | N/A | 63.8 |
| CY1300 + TiO ₂ | Micro (1500) | 1 | 76.1 |
| CY1300 + TiO ₂ | Micro (1500) | 10 | 73.9 |
| CY1300 + TiO ₂ | Micro (1500) | 50 | 79.9 |
| CY1300 + TiO ₂ | Nano (38) | 1 | 62.9 |
| CY1300 + TiO ₂ | Nano (38) | 10 | 52.4 |
| CY1300 + TiO ₂ | Nano (38) | 50 | 62.1 |

Table 1 Glass transition of nano- and micro-filled TiO₂

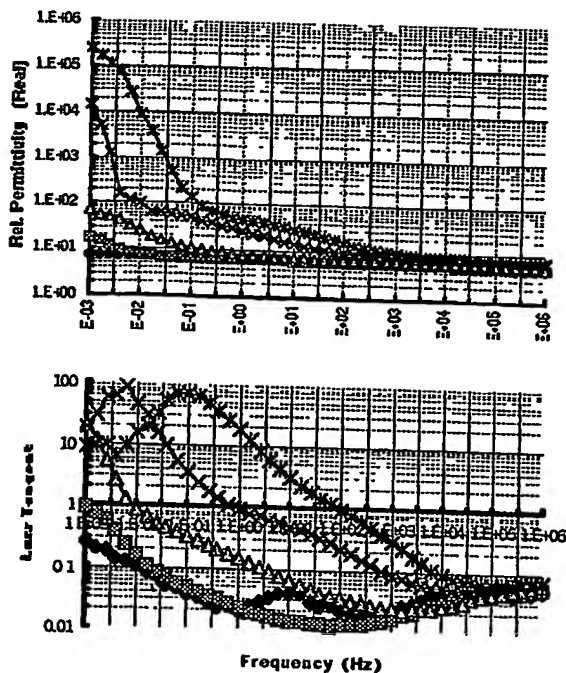


Figure 1. Permittivity and loss tangent for micro-filled epoxy. Temps. (bottom to top) 293, 318, 343, 368, 393 K

and with a glass transition below 100 °C. Test samples were formed by molding between polished surfaces held apart by spacers in the manner previously described[7]. The molded films ranged in thickness between 500 and 750 μm . The weighed resin and hardener were degassed at 35 °C and the relevant dried particulate fill was incorporated into the resin by mechanical stirring. Due to their small size, surface interactions for nanoparticles, such as hydrogen bonding, become magnified. This means that the particles tend to agglomerate and dispersion in resins is quite difficult, even in polymers that should be relatively compatible. Hence, in the case of nano-particles, large shear forces are needed in the mixing process to obviate unwanted clustering of the particles. For most electrical characterization, the cast film was provided with evaporated 100 nm aluminium electrodes.

Differential Scanning Calorimetry (DSC)

A Stanton Redcroft DSC 1500 calorimeter was used to thermally characterize the materials. Results on the determination of glass transition temperatures are provided in Table 1 for post-cured samples from which it is evident that the nano-material reduces T_g in contrast to the larger size particles that have the opposite effect.

This suggests that particles of nanometric dimensions behave in a similar way to infiltrated plasticizers[8], rather than as "foreign" materials creating a macroscopic interface.

Dielectric Spectroscopy

Some insight into the way that the incorporation of materials on nanometric dimensions affect the dielectric

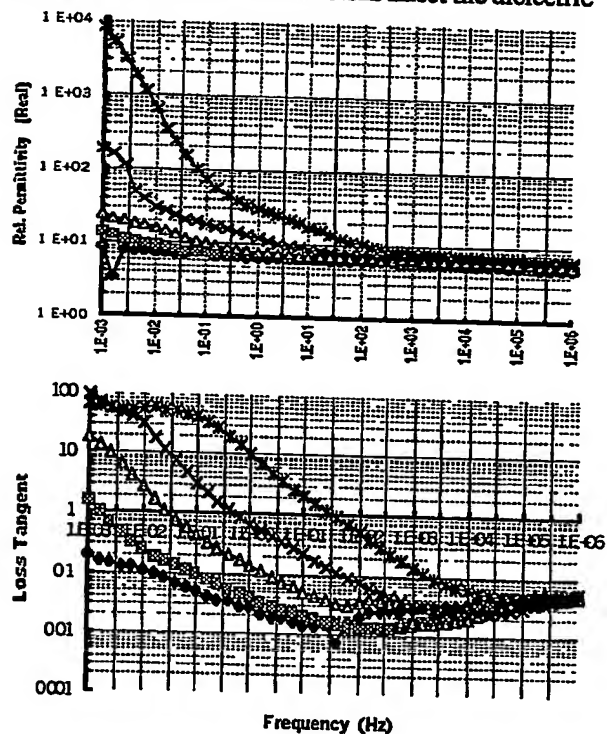


Figure 2. As Figure 1, but for nano-filled material

properties may be obtained by examining the variation of the real and imaginary components of relative permittivity as a function of temperature and frequency. This has been done for the TiO_2 material using a Solartron H.F. frequency response analyzer (Type 1255) in combination with a Solartron Dielectric Interface, Type 1296. Examples for the micro- and nano-filled materials are shown in Figs. 1 and 2 respectively. At a nominal 10% (weight percent) particulate loading, the spectra of the resin when filled with particles of micron size (1.5 μm) are virtually indistinguishable from the base resin. This suggests that the low frequency process is probably associated with charges at the electrodes and not due to particulates in the bulk. With the filler replaced with 10% of nanometric size TiO_2 (38 nm

average diameter measured by TEM), the main differences seen relate to a marked modification of the process seen in the base resin at low frequencies and high temperatures. For the nanometric material the process exhibits a flat $\tan \delta$ response at low frequencies in marked contrast to the micron-sized filler. This suggests that a percolation conduction process is operative. In the presence of the nano-filler, the mid frequency dispersion is noticeably reduced. The nano materials are clearly inhibiting motion (see PEA results below). The mid-frequency process shows a small change in estimated activation energy from 1.7 eV to 1.4 eV. The magnitude of this process is reduced in the case of nanoparticles since the side chains responsible for the mid-frequency dispersion bind to the particle surface.

Reduction of the particulate loading from 10 to 1% (by weight) did not have any very obvious fundamental changes, but the nano-filled material then does start to exhibit a low frequency response more typical of the base resin and micro-filled material, suggesting that changes engineered by the nanomaterials do require loadings greater than a few percent.

Space charge assessment

In order to determine whether nanomaterials function cooperatively as opposed to providing sites for interfacial polarization, a Pulse ElectroAcoustic (PEA) study has also been conducted to assess the field distortions in the bulk. The method has been described elsewhere[9]. The initial distribution of stress shows

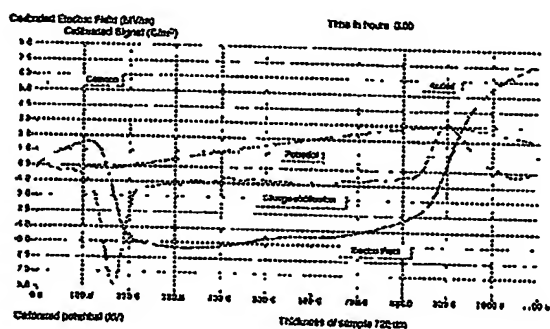


Figure 3 Initial distribution of electric field. Electroacoustic study of nano-filled material

little deviation from the nominal 4.3 kVmm^{-1} uniform level across the bulk (see Figure 3). However, characteristic results are shown in Figs. 4 and 5 for the micro- and nano-materials (10% loading) respectively

after several hours of stressing. These plots show the charge, potential and field distributions, for a 3 kV steady DC field applied. The $1.5\mu\text{m}$ filler generates

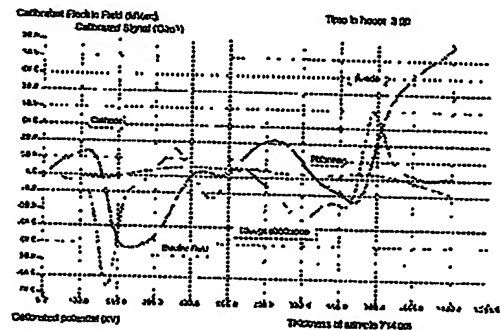


Figure 4. Pulsed electroacoustic study of micron-sized filler.

substantial internal charge, in marked contrast to the nano-material which behaves in a similar way to the

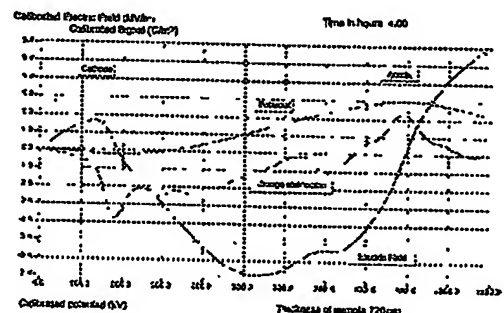


Figure 5. Pulsed electroacoustic study of nano-sized filler

base resin. Fig 4 shows several distinctive features:

- (a) heterocharge accumulation of both signs leading to steep internal charge gradients
- (b) a cathode field augmented to over 40 kVmm^{-1} (10x the nominal value)
- (c) field reversal yielding a point of zero stress which will greatly complicate charge transport.

Transient studies (not shown here) indicate that subsequent increase of applied voltage increases the size of the charge peaks with little change to the complex internal distribution. The stable stationary positioning of these peaks may be due the interaction of space charge with local polarization to create a self-compensating situation.

Appraisal

Very marked differences in charge accumulation are seen in filled materials depending on whether the filler has micron or nanometric dimensions. Furthermore, the characteristics suggest that, for the micron-sized filler, carriers are blocked at the anode yielding a heterocharge situation, and giving rise to the large anomalous field distortions seen in Figure 4. This behavior clearly has substantial implications for the subsequent migration of charges and probably accounts for the fact that temporal studies (not given here) show that the image charge in the cathode at first decreases and then recovers. Again in contrast to the micro-filled material, the decay of charge in the nano-filled TiO_2 is very rapid; with insignificant homocharge remaining after just 2 minutes. Although there is some injection of negative charge at the cathode, the nano-filled material is characterized by much less transport perhaps brought about by the larger density of shallower traps.

The PEA results taken in conjunction with the Dielectric Spectroscopy and DSC studies suggest that significant interfacial polarization is implied for conventional fillers which is mitigated in the case of particulates of nanometric size, where a short-range highly immobilized layer develops near the surface of the nanofiller (1-2 nm). This bound layer, however, influences a much larger region surrounding the particle in which conformational behavior and chain kinetics are significantly altered. This *interaction zone* is responsible for the material property modifications especially as the curvature of the particles approaches the chain conformation length of the polymer. Evidence suggests that the local chain conformation and configuration play major roles in determining the interactions of a polymer with nanofillers[10], as is evidenced here by the DSC results of Table 1. The polymer binding to the nanoparticles replaces some of the cross-linking and thus loosens the structure. In contrast, the micron scale case produces significant Maxwell-Wagner polarization giving rise to the characteristics of Fig.4.

In the case of nanofillers, there is evidence that a grafted layer is formed by the absorption of end-functionalised polymers onto the surface especially when the functional groups are distributed uniformly along the polymer backbone. Hence the local chain conformation is critical to determining the way in which bonding takes place (and thus the cohesive energy density). The defective nature of nanoscale particles can be expected to enhance the bonding if chemical coupling agents (CVD coatings on nanoparticles or triblock copolymers) are employed.

The large interaction zone in nanofilled polymers with reduced mobility (free volume) should be accompanied by a significant change in electrical properties. Studies of electrical behavior thus provide an opportunity both for a fundamental study of this interaction zone, and also an opportunity for optimizing performance for specific and critical applications.

The finding that conventional fillers are accompanied by substantial bulk charge accumulation is clearly a factor in the common experience of the lower electric strengths exhibited for filled materials. The mitigating effects of nanoparticles provides encouragement that nanocomposites can be engineered with strengths that are commensurate with the base polymer. Such studies are ongoing.

Acknowledgements

The authors are indebted to the UK EPSRC under whose auspices this study has been started, and for the loan of the DSC equipment. Thanks are also due to Rensselaer for sabbatical leave, and the provision of nanometric powders through the Nanotechnology Center.

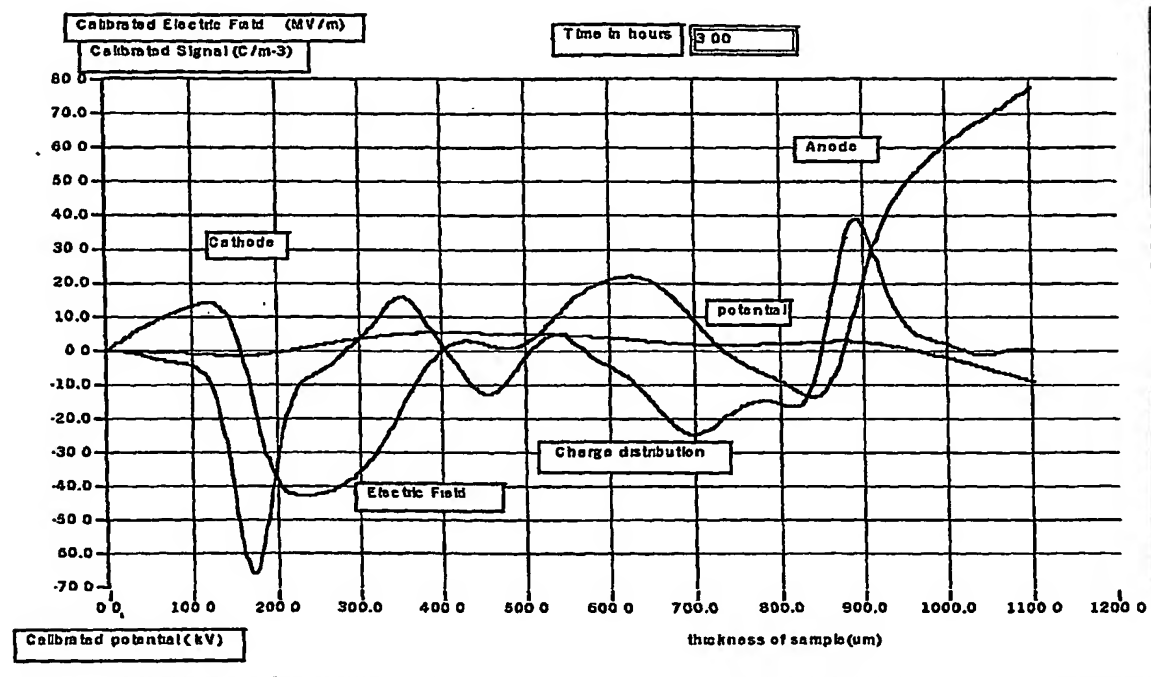
References

- [1] Lewis T.J., "Nanometric Dielectrics", IEEE Trans on Dielect. And Elect. Ins., Vol.1, pp 812-25, 1994
- [2] Frechette M.F. et al., "Introductory remarks on NanoDielectrics", Ann. Rep. Conf. On Elect. Ins. And Dielect. Phen., IEEE, pp 92-99, 2001
- [3] Sabuni H. and Nelson J.K., "Factors determining the electric strength of polymeric dielectrics", J. Mats Sci., Vol. 11, p1574, 1976
- [4] Nelson J.K., "Breakdown strength of solids", in *Engineering Dielectrics*, Vol. 2A, ASTM, 1993
- [5] Crine J-P., Vijn A.K., "Molecular approach to the physico-chemical factors in the electrical breakdown of polymers", Appl. Phys. Comm., Vol.5, p 139-63, 1985
- [6] Dissado L.A. and Fothergill J.C., "Electrical degradation and breakdown in polymers", Peter Peregrinus, 1992
- [7] Griseri V., "The effects of high electric fields on an epoxy resin", Ph.D. Thesis, University of Leicester, 2000
- [8] Sabuni M.H. and Nelson J.K., "The effects of plasticizer on the electric strength of polystyrene", J. Mats. Sci., Vol.14, pp 2791-96, 1979
- [9] Alison J., "A High Field Pulsed Electro-Acoustic Apparatus for Space Charge and External Circuit Current Measurement within Solid Dielectrics", Meas. Sci Technol., Vol. 9, pp 1737-50, 1998
- [10] Schmidt-Rohr K. and Spiess H.W., "Nature of non-exponential loss of correlation above the glass transition investigated by multidimensional NMR", Phys. Rev Lett., Vol. 66, p 3020, 1991

ATTACHMENT 1.

SAMPLE Ti/006

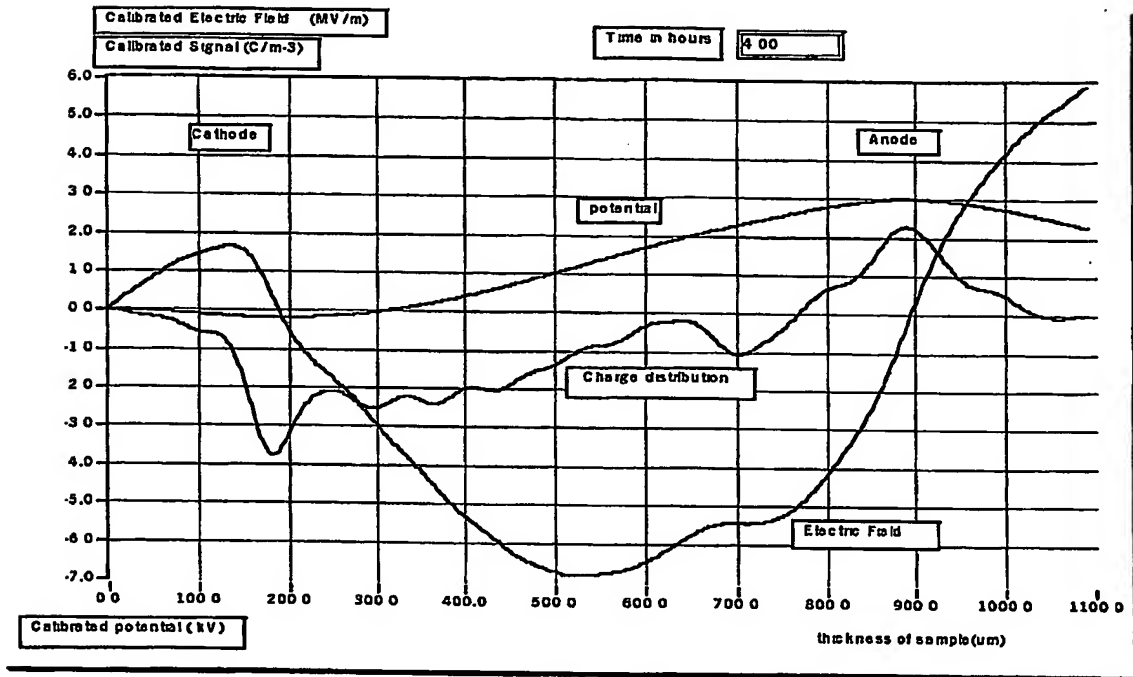
10% TiO₂ 1.5 μm with no electrodes: 3kV for 4 Hours



| | |
|------------------------|--|
| Thickness: | 714 μm |
| Interrogating pulse | 5 ns +400V, 100 pps |
| Scope: | 5 ns, 5 mV, 1.718 μs delay |
| Acoustic Delay: | 259 ns |
| Speed of sound: | 2757 ms^{-1} |
| Sample Rate: | 1.25 GSs ⁻¹ |
| Integration Range: | 397 (876 thickness) |
| Relative permittivity: | 6.0 at 100 Hz |
| Voltage bias: | ON 3 kV DC |
| Data column: | 0 |
| Filter: | 6.0 |
| Data file: | Ti006-5.txt |
| Timebase file: | time.txt |
| Reference file: | irf40new.txt (Modified PMMA reference) |
| Output file: | Ti006-5.doc |

SAMPLE Ti/005R (REPEAT MEASUREMENT)

10% TiO₂ 38 nm with no electrodes: 3 kV for 4 hours



| | |
|------------------------|--|
| Thickness: | 726 μm |
| Interrogating pulse | 5 ns +400V |
| Scope: | 5 ns, 5 mV, 1.724 μs delay |
| Acoustic Delay: | 255 ns |
| Speed of sound: | 2726 ms ⁻¹ |
| Sample Rate: | 1.25 GSs ⁻¹ |
| Integration Range: | 410 (894 thickness) |
| Relative permittivity: | 6.2 |
| Voltage bias: | ON 3 kV DC |
| Data column: | 0 |
| Filter: | 6.0 |
| Data file: | Ti005R-5.txt |
| Timebase file: | time.txt |
| Reference file: | irf40new.txt (Modified PMMA reference) |
| Output file: | Ti005R-5.doc |

**This Page is Inserted by IFW Indexing and Scanning
Operations and is not part of the Official Record**

BEST AVAILABLE IMAGES

Defective images within this document are accurate representations of the original documents submitted by the applicant.

Defects in the images include but are not limited to the items checked:

- ☐ BLACK BORDERS
- ☐ IMAGE CUT OFF AT TOP, BOTTOM OR SIDES
- ☐ FADED TEXT OR DRAWING
- ☒ BLURRED OR ILLEGIBLE TEXT OR DRAWING
- ☐ SKEWED/SLANTED IMAGES
- ☐ COLOR OR BLACK AND WHITE PHOTOGRAPHS
- ☐ GRAY SCALE DOCUMENTS
- ☒ LINES OR MARKS ON ORIGINAL DOCUMENT
- ☐ REFERENCE(S) OR EXHIBIT(S) SUBMITTED ARE POOR QUALITY
- ☐ OTHER: _____

IMAGES ARE BEST AVAILABLE COPY.

As rescanning these documents will not correct the image problems checked, please do not report these problems to the IFW Image Problem Mailbox.